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(54) **Binaphthalene derivatives for organic electro-luminescent devices**

(57) The present invention relates to an organic light emitting device (OLED) in which a binaphthalene or biquinoline derivative is used as the emissive layer and/or one or more of the charge transport layers, or as a host or dopant material for one or more of such layers.

Description

BACKGROUND OF THE INVENTIONField of the Invention

[0001] The present invention relates to an organic light emitting device (OLED) in which a binaphthalene derivative is used as the emissive layer and/or one or more of the charge transport layers, or as a host or dopant material for one or more of such layers.

Description of the Related Art

[0002] Significant efforts have been expended in developing suitable materials for use in organic light emitting devices (OLEDs). Such devices are commercially attractive because they offer the promise of low-cost fabrication of high-density pixelated displays exhibiting bright electroluminescence with long life times and a wide range of colors.

[0003] A typical OLED is fabricated by sandwiching an emissive layer between an anode and a cathode. Improved performance can be obtained by the provision of additional layers around the emissive layers so as to provide charge transport capabilities, such as an electron transport layer or a hole transport layer. Some of these layers can be combined. For example, a double-layered structure is fabricated from a combined hole-injecting and transporting layer together with a combined electron-transporting and light-emitting layer. Likewise, a triple-layered structure is composed of a hole-injecting and transporting layer, a light-emitting layer, and an electron-injecting and transporting layer.

[0004] In addition, it is possible to form these layers from a host material doped with another material designed to achieve the desired effect of the layer (that is, to achieve a hole transport effect, an electron transport effect, or an emissive effect).

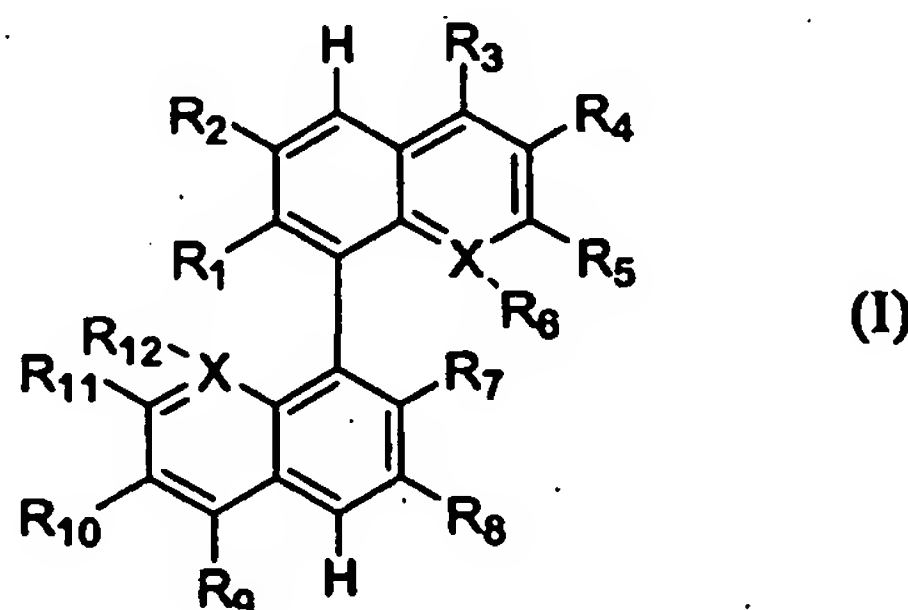
[0005] Because of consumer expectations of good efficiency, long life time and pure color, a need exists for development of suitable materials for the various layers.

SUMMARY OF THE INVENTION

[0006] It is an object of the invention to provide an improved OLED in which a binaphthalene derivative is used as the emissive layer and/or one or more of the charge transport layers, or as a host or dopant material for one or more of such layers.

[0007] 1, 1' binaphthalene derivatives represent an important class of chiral auxiliaries. The large dihedral angle between the naphthyl groups ensures a high degree of stereoselectivity and the resulting non co-planar binaphthyl unit prevents close molecular packing. As a result, when incorporated into OLEDs, especially in the emissive layer, a high photo-luminescent efficiency and a possible high electro-luminescent efficiency can be achieved.

[0008] Thus, in one aspect, the invention is an OLED in which an emissive layer is sandwiched between at least a cathode and an anode, and in which the emissive layer includes a binaphthalene or biquinoline derivative expressed according to the following general formula (I):



wherein each of R_1 to R_{12} independently represents a hydrogen atom, an alkyl group, an alkoxy group, a vinyl group, an amine group, an aromatic compound, or a hetero-aromatic compound, and the 4 and 4' positions on the naphthalene or quinoline rings are unsubstituted as shown. X can represent either carbon or nitrogen atoms, however, if X represents nitrogen, then R_6 and R_{12} are absent.

[0009] The compound expressed according to formula (I) can be used directly as the emissive layer, or can be used as a host material for an emissive dopant in a case where the emissive layer comprises a binaphthalene or biquinolne derivative host plus an emissive dopant. The emissive dopant can also be a compound expressed according to formula (I) as specified above.

5 [0010] When the compound expressed according to formula (I) is a chiral molecule, and when the optically active enantiomers of the compound are isolated into separate R and S enantiomers, the light produced in the emissive layer by the resolved binaphthalene derivative is polarized.

[0011] In a further aspect, the invention is an OLED having an emissive layer sandwiched between at least one charge transport layer and an anode and a cathode. The charge transport layer can be either an electron transport layer or a hole transport layer, or both. According to this aspect of the invention, the charge transport layer includes a binaphthalene or biquinolne derivative according to the above general formula (I) as specified above.

[0012] According to this aspect, the compound expressed according to formula (I) can be used directly as the charge transport layer, or can form a charge transport host material in a case where the charge transport layer comprises a host material plus a charge transport dopant.

15 [0013] This brief summary has been provided so that the nature of the invention may be understood quickly. A more complete understanding of the invention can be obtained by reference to the following detailed description of the preferred embodiment thereof in connection with the attached drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

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[0014] Figure 1 is a differential scanning calorimetry (DSC) curve of Compound 2.

[0015] Figure 2 is a PL spectrum of Compound 4 in CHCl_3 .

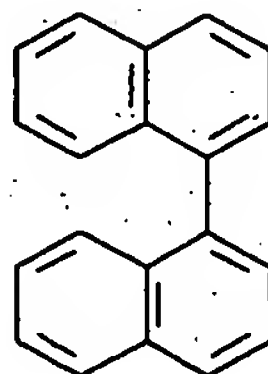
[0016] Figure 3 is a schematic cross-sectional view of an OLED according to the invention.

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DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0017] Binaphthalene has the following structure:

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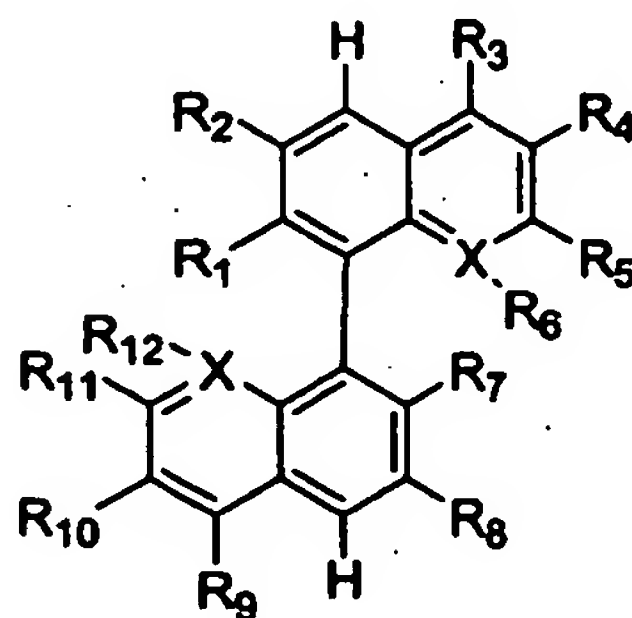


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[0018] The present invention relates to organic light-emitting devices containing binaphthalene derivatives having the following general structure:

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[0019] In the above formula (I), R_1 through R_{12} can be substituted or unsubstituted independently, depending upon the starting materials used in the synthesis, or to achieve desired properties. Substituents may include, without limi-

tation, a hydrogen atom, an alkyl group, an alkoxy group, a vinyl group, an amine group, an aromatic compound, or a hetero-aromatic compound. Preferably, R_1 through R_{12} are emissive, and/or charge-transporting groups for organic light-emitting device (OLED) applications.

[0020] As shown, the 4 and 4' positions on the naphthalene rings are unsubstituted. Additionally, in the above formula (I), X represents either a carbon or a nitrogen atom, however, if X represents a nitrogen atom, then R_6 and R_{12} are absent.

[0021] The binaphthylene and biquinoline derivatives used in the present invention are small molecules as opposed to large polymers.

[0022] In a preferred embodiment of the present invention, the optically active enantiomers of the compound are isolated into separate R and S enantiomers. When the binaphthalene or biquinoline derivative is resolved and used as an emissive layer in an OLED, the light emitted will be polarized.

[0023] According to another preferred embodiment, the binaphthylene or biquinoline derivative is substituted at the 2 and 2' positions, giving the chemical structure a twisted conformation. In this embodiment, R_1 and R_7 , in Formula (I) represent a substituent group other than hydrogen. More preferably, R_1 and R_7 represent substituents which impart the compound with emissive, hole transport or electron transport properties. Arylamine groups are examples of suitable hole transport groups. Substituents which may be used to impart electron transport properties include, but are not limited to, oxadiazoles, thiadiazoles or triazoles, diaryl sulfones, aryl sulfoxide, fluorinated aryls, biphenyls, diaryl phosphine oxides and benzophenones. Aryl-substituted oxadiazole, thiadiazole or triazole are particularly preferred as electron transport substituents.

[0024] Figure 1 schematically depicts an OLED according to the invention, including transparent substrate 1, anode 2 adjacent the substrate, hole transport layer 3 adjacent the anode, emissive layer 4, electron transport layer 5, and cathode 6. Each of these layers may itself comprise multiple layers of material having similar composition or function.

[0025] A binaphthylene or biquinoline of formula (I) can be used in an OLED in a number of ways. In one aspect, the binaphthalene or biquinoline derivative is included in the emissive layer of an OLED, in which the emissive layer is sandwiched between at least one cathode and one anode. Here, the binaphthalene or biquinoline derivative can be the emissive layer host, the emissive dopant, or both.

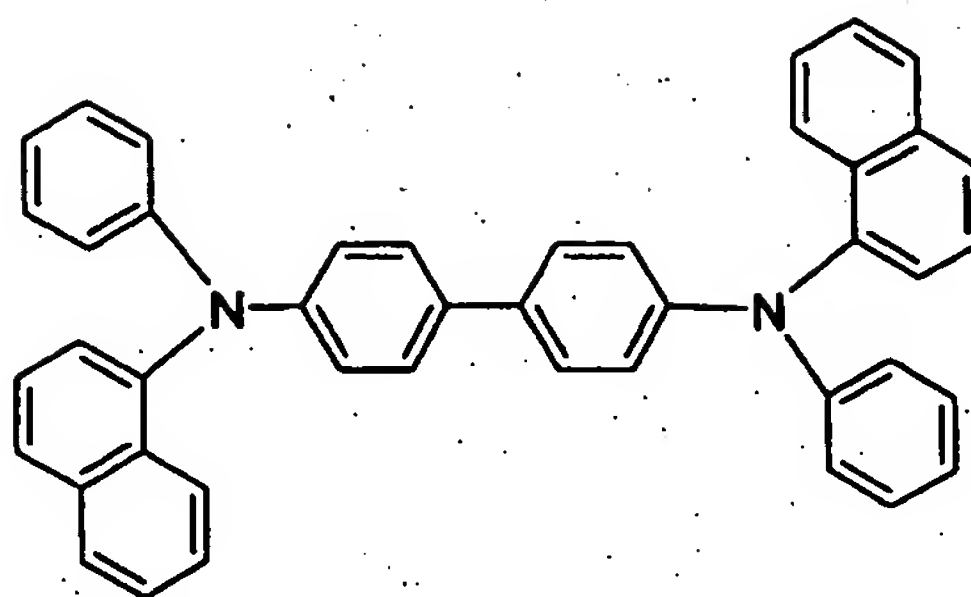
[0026] In addition to the emissive layer, another OLED of the present invention has a charge-transport layer sandwiched between a cathode and an anode, where the charge-transport layer includes a binaphthalene or biquinoline derivative conforming to formula (I) described above. The derivative can be a component of the electron-transport layer, the hole-transport layer or both. As a component of these layers, the binaphthalene or biquinoline derivative can serve as either host or charge transport dopant.

[0027] Binaphthylene materials suitable for use with the invention or as starting materials for preparing derivatives according to the invention, can be made using methods known in the art. These methods include, but are not limited to Suzuki coupling reactions and palladium(0)-catalyzed amination reactions. See *Accounts of Chemical Research*, Vol. 31, p. 805 (1998); *Angewandte Chemie International Edition English*, Vol. 37, p. 2047 (1998); *Chemical Reviews*, Vol. 95, No. 7, pp. 2457-2483 (1995); and *Tetrahedron Letters*, Vol. 39, p. 617 (1998), incorporated herein by reference.

[0028] An advantage of the disclosed binaphthylene or biquinoline derivative materials for use in OLEDs according to the invention is that they prevent close molecular packing. This advantage is due to the large dihedral angle between the two naphthyl groups. When close molecular packing is prevented, the result in an OLED is high photo and electroluminescent efficiency.

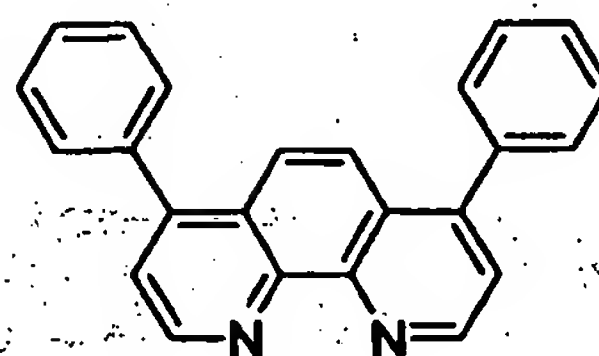
[0029] OLEDs of the present invention can be fabricated by first obtaining a clean substrate with an ITO patterned thereon. Then, the substrate is treated with O_2 plasma for 1 to 5 minutes. Once treated, the substrate is placed in a thermal evaporator and the pressure is pumped down below 6×10^{-6} torr. An emissive layer, either with or without a dopant, is then evaporated onto the substrate. Optionally, a hole transport layer and/or an electron transport layer are also evaporated onto the substrate. Finally, another electrode layer is deposited thereon.

[0030] A preferred hole transporting material is 1,4-bis[(1-naphthylphenyl)-amino]biphenyl (NPD) and has the following structure:



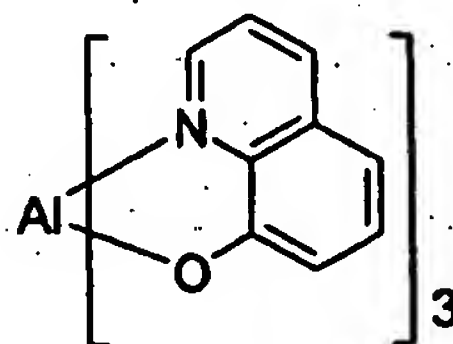
1,4-bis[(1-naphthyl)amino]biphenyl (NPD)

[0031] A preferred electron transport material is 4,7-diphenyl-1,10-phenanthroline (Bphen) and has the following structure:



4,7-diphenyl-1,10-phenanthroline (Bphen)

[0032] A suitable emissive material which may be used in the present invention is aluminum tris(8-hydroxyquinoline) (AlQ3) having the following structure:

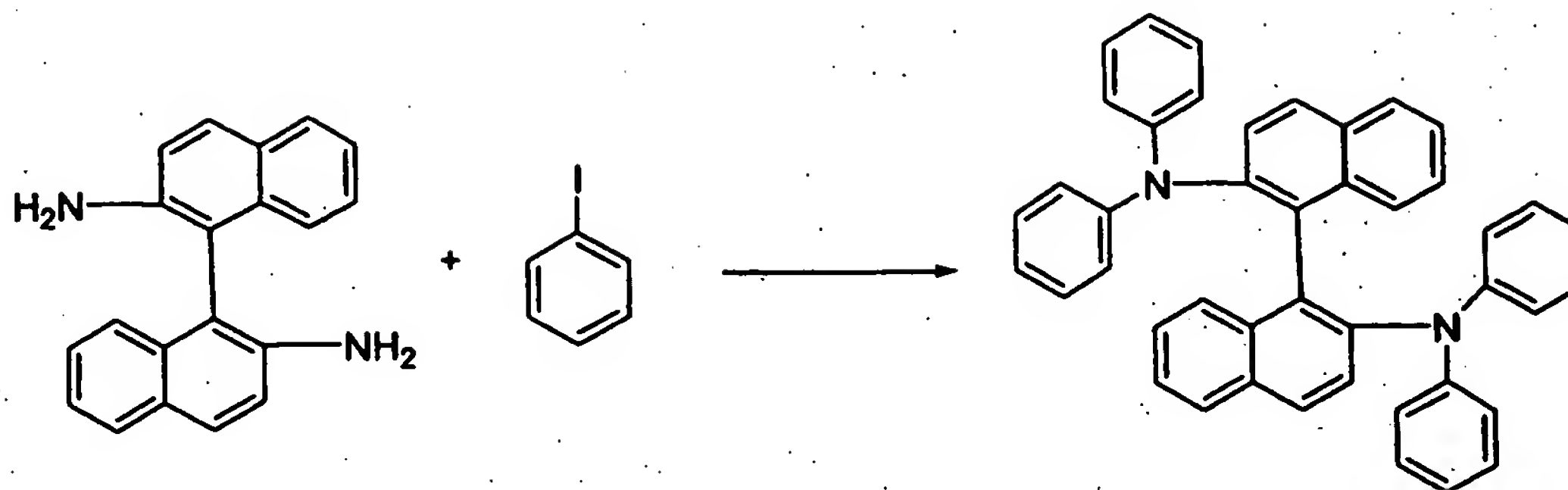


AlQ₃ (aluminum tris(8-hydroxyquinoline))

[0033] A number of binaphthalene derivatives have been synthesized to test their suitability for use in OLEDs.

EXAMPLE 1

[0034]

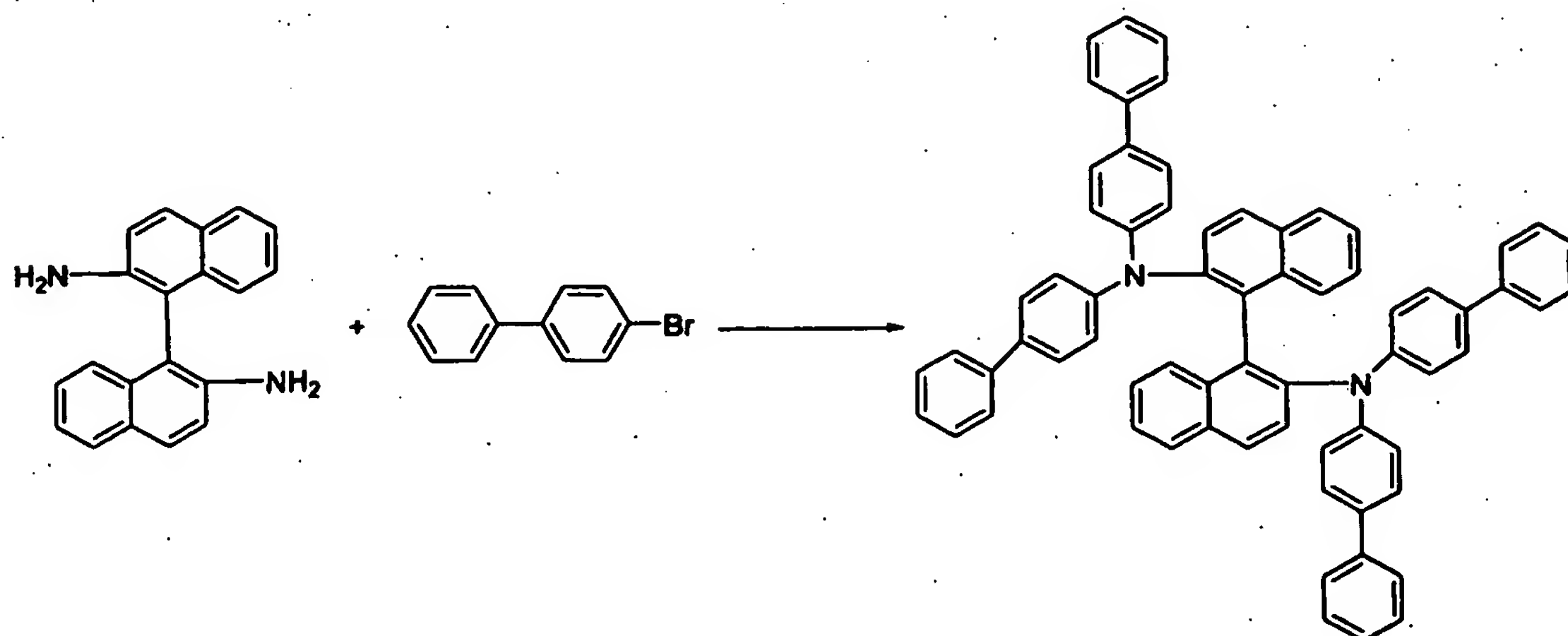


Compound (1)

[0035] Compound (1) was synthesized using palladium(0)-catalyzed amination reactions as described in references cited above. A round-flask was charged with 2,2'-diamino-1,1'-binaphthyl (0.568 g, 2 mmol), iodobenzene (1.14 mL, 10 mmol), NaOt-Bu (0.96 g, 10 mmol), bis(tri-*t*-butylphosphine)palladium(0) (50 mg) and 1,4-dioxane (25 mL). The mixture was stirred at 80 °C under N₂ for two days. After cooling down, the mixture was poured into water. The product was then extracted with toluene, washed with water and dried over MgSO₄. After filtration and concentration, it was purified through column chromatography (Silica gel, mixtures of CH₂Cl₂/Hexanes). A thermogravimetric analysis (TGA) scan indicated a thermal decomposition temperature of 305 °C. Compound (1) can be fabricated into an OLED using the above generalized procedure.

EXAMPLE 2

[0036]



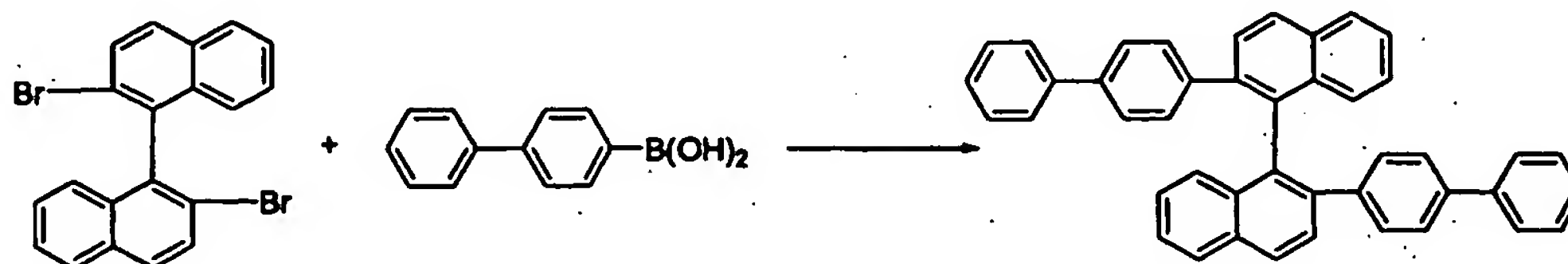
Compound (2)

[0037] Compound (2) was synthesized in a way similar to Compound 1, as described above. The difference being

that 4-bromobiphenyl was used in place of iodobenzene. Differential scanning calorimetry (DSC) showed a very high melting point of 337°C, while TGA indicated a high decomposition temperature of 440°C. The results of the DSC scan are shown in Figure 1. Compound 2 has a HOMO value of -5.58 eV, and a LUMO of -2.48 eV. Compound (2) can be fabricated into an OLED using the above generalized procedure.

EXAMPLE 3

[0038]

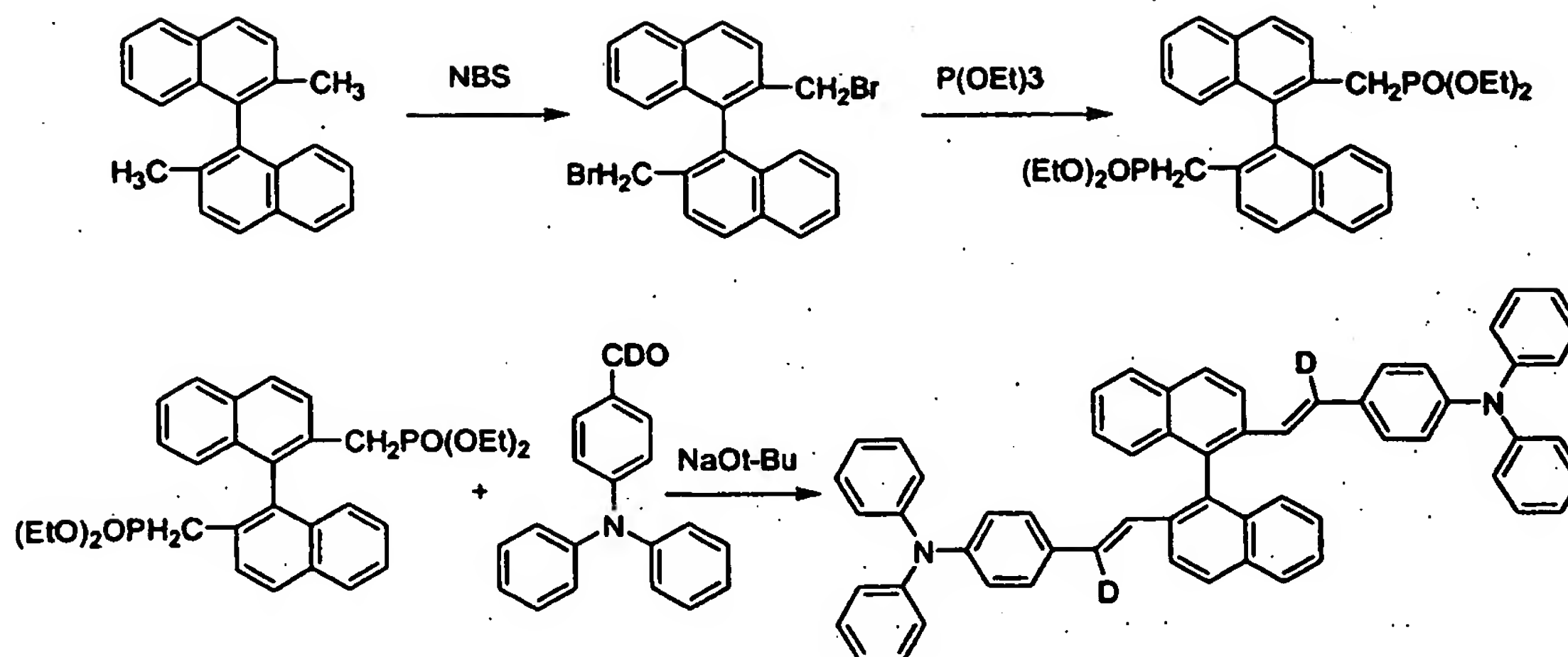


Compound (3)

[0039] Compound (3) was synthesized using Suzuki coupling reactions and palladium(0)-catalyzed amination reaction, described in the references cited above. A round-flask was charged with 2,2'-dibromo-1,1'-binaphthyl (0.824 g, 2 mmol), 4-biphenylboronic acid (1.6 g, 8 mmol), $\text{Pd}(\text{P}-t\text{-Bu}_3)_2$ (80 mg), NaOt-Bu (0.96 g, 10 mmol), and 30 mL of dry o-xylene. The mixture was stirred at 110 °C under N_2 for two days. After cooling down, the mixture was poured into water. The product was then extracted with toluene, washed with water, and dried over MgSO_4 . After filtration and concentration, it was purified through column chromatography (Silica gel, mixtures of CH_2Cl_2 /Hexanes), and recrystallized from toluene/hexane. A DSC scan indicated the compound has a melting temperature of 272 °C. Compound (3) can be fabricated into an OLED using the above generalized procedure.

EXAMPLE 4

[0040]



Compound (4)

[0041] Compound (4) was synthesized via a few-step reactions. 2,2'-dibromomethyl-1,1'-binaphthyl was obtained by reaction of NBS with 2,2'-dimethyl-1,1'-binaphthyl according to a procedure disclosed in *Synthesis*, pp. 317-320 (1985) incorporated herein by reference. Refluxing of 2,2'-dibromomethyl-1,1'-binaphthyl in an excess of triethylphosphite afforded the ester compound. The deuterated 4-aldehydetriphenylamine was obtained by a reaction of triphenylamine (1 equiv.) and deuterated DMF- d_7 (1.4 equiv.) in the presence of $POCl_3$ (1.2 equiv.) according to a similar reference procedure disclosed in *Chemistry of Materials*, Vol. 11, pp. 1568-1575 (1999). Finally, a round-flask was charged with a 1,1'-binaphthyl ester compound (0.77 g, 1.39 mmol), deuterated 4-aldehydetriphenylamine (0.8 g, 2.92 mmol), $NaOt-Bu$ (0.4 g, 4.17 mmol), and 10 mL of dry DMF. The mixture was stirred at room temperature under N_2 for two days. The mixture was poured into 100 mL of water. The precipitate was filtrated and washed with water and methanol. The finished product was purified through column chromatography (Silica gel, mixtures of CH_2Cl_2 /Hexanes). Compound (4) has a T_g at 122 °C by DSC and shows a strong blue emission as shown in Figure 2. This compound can be fabricated into an OLED using the above generalized procedure

EXAMPLE 5

[0042] An OLED was made in accordance with the procedures disclosed above. The device was constructed with a ITO anode with NPD deposited thereon to act as a hole transporter. Bphen was used as an electron transporter while a lithium aluminum alloy was used as the cathode. In this device, an emissive layer was also present. Compound (3) was the host, while Compound (4) was the dopant.

[0043] The driving voltage was applied to the obtained device by connecting the ITO electrode to a positive electrode (+) and the cathode of Al to a negative electrode(-) to test emission of light, and a uniform light of blue color was obtained. The density of the electric current produced by the device was 100 mA/cm² at 7 V; the luminescence of the device was measured at 1350 cd/m² at 7 V; the maximum external quantum efficiency of the OLED was 1.2 % and the maximum external power efficiency 1.0 lm/W. CIE color coordinates were (0.19, 0.22).

EXAMPLE 6

[0044] An OLED was fabricated in accordance with the procedures disclosed above, where Compound (1) was a hole transporter and AlQ₃ was a green emitter and electron transporter. Upon applying a positive bias, the device emitted green light, peaking at 520 nm, typical of AlQ₃ emission. The luminescence of the device was 10,000 cd/m² at 14 V.

EXAMPLE 7

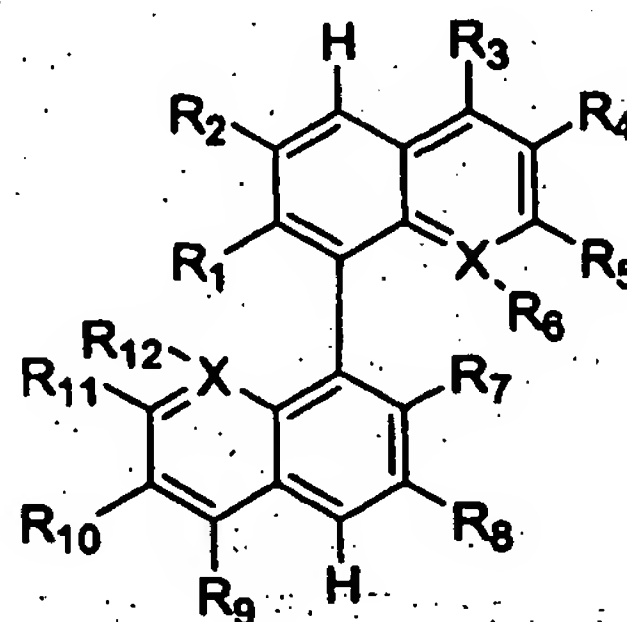
[0045] Another OLED was fabricated in accordance with the procedures disclosed above, where Compound (2) was a hole transporter, AlQ₃ was a green emitter and electron transporter. Upon applying a positive bias, the device emitted green light, peaking at 520 nm, which is typical of an AlQ₃ emission. The luminescence of this device was measured at 12,000 cd/m² at 11 V.

[0046] The foregoing examples are illustrative only and are not to be deemed limiting of the invention, which is defined by the following claims and is understood to include such obvious variations and modifications as would be obvious to those of ordinary skill in the art.

Claims

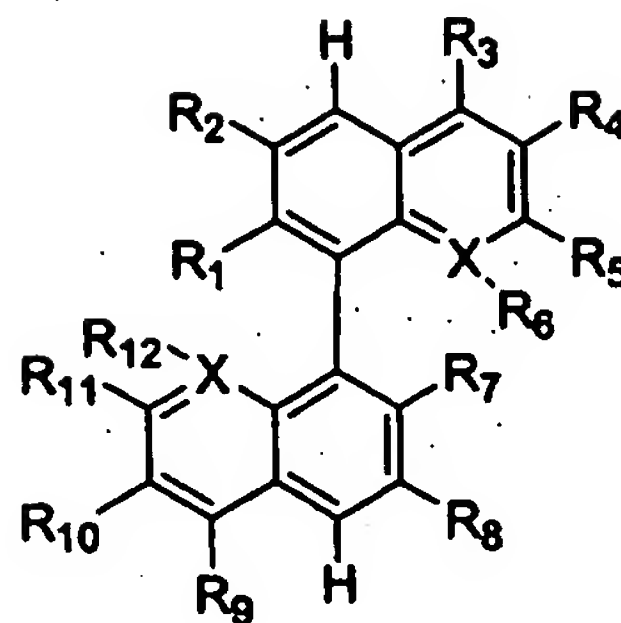
1. An organic light emitting device comprising:

an emissive layer sandwiched between at least a cathode and an anode, wherein the emissive layer includes a first binaphthalene or biquinoline derivative expressed according to the following general formula:



wherein each of R₁ to R₁₂ independently represents a hydrogen atom, an alkyl group, an alkoxy group, a vinyl group, an amine group, an aromatic compound, or a hetero-aromatic compound;
 wherein the 4 and 4' positions on the naphthalene rings are unsubstituted;
 wherein X can represent either carbon or nitrogen atoms; and
 wherein if X represents nitrogen, then R₆ and R₁₂ are absent.

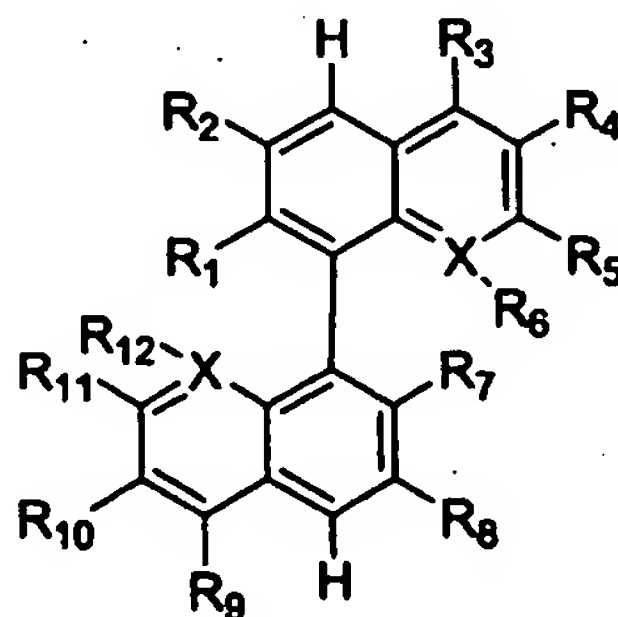
2. An organic light emitting device according to Claim 1, wherein said emissive layer is comprised of a functional light-emitting layer of the first binaphthalene derivative.
3. An organic light emitting device according to Claim 1, wherein said emissive layer is comprised of a host material of the first binaphthalene derivative, together with a functional light emitting dopant.
4. An organic light emitting device according to Claim 3, wherein said functional light emitting dopant is a second binaphthalene or biquinoline derivative expressed according to the following general formula:



wherein each of R_1 to R_{12} independently represent a hydrogen atom, an alkyl group, an alkoxy group, a vinyl group, an amine group, an aromatic compound, or a hetero-aromatic compound;
 wherein the 4 and 4' positions on the naphthalene or quinoline rings are unsubstituted;
 wherein X can represent either carbon or nitrogen atoms; and
 wherein if X represents nitrogen, then R_6 and R_{12} are absent.

5. An organic light emitting device according to Claim 1, wherein said first binaphthalene derivative is comprised substantially of one of the optically active enantiomers of the first binaphthalene derivative.
6. An organic light emitting device according to Claim 1, wherein the first binaphthalene derivative is substituted at the 2 and 2' positions.
7. An organic light emitting device according to Claim 1, wherein each of R_1 to R_{12} independently represent a hydrogen atom or an emissive group.
8. An organic light emitting device comprising:

an emissive layer sandwiched between at least one charge transport layer and an anode and a cathode;
 wherein the charge transport layer includes a binaphthalene derivative expressed according to the following general formula:



wherein each of R_1 to R_{12} independently represent a hydrogen atom, an alkyl group, an alkoxy group, a vinyl group, an amine group, an aromatic compound, or a hetero-aromatic compound;
 wherein the 4 and 4' positions on the naphthalene or quinoline rings are unsubstituted;
 wherein X can represent either carbon or nitrogen atoms; and
 wherein if X represents nitrogen, then R_6 and R_{12} are absent.

9. An organic light emitting device according to Claim 8, wherein said charge layer is comprised of a functional charge transport layer of the binaphthalene derivative.

10. An organic light emitting device according to Claim 8, wherein said charge transport layer is comprised of a host material of the binaphthalene derivative, together with a functional charge transport dopant.

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11. An organic light emitting device according to Claim 8, wherein said charge transport layer is an electron transport layer.

12. An organic light emitting device according to Claim 8, wherein said charge transport layer is a hole transport layer.

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13. An organic light emitting device according to Claim 8, wherein the first binaphthalene derivative is substituted at the 2 and 2' positions.

14. An organic light emitting device according to Claim 8, wherein each of R_1 to R_{12} independently represent a hydrogen atom, or a charge transport group.

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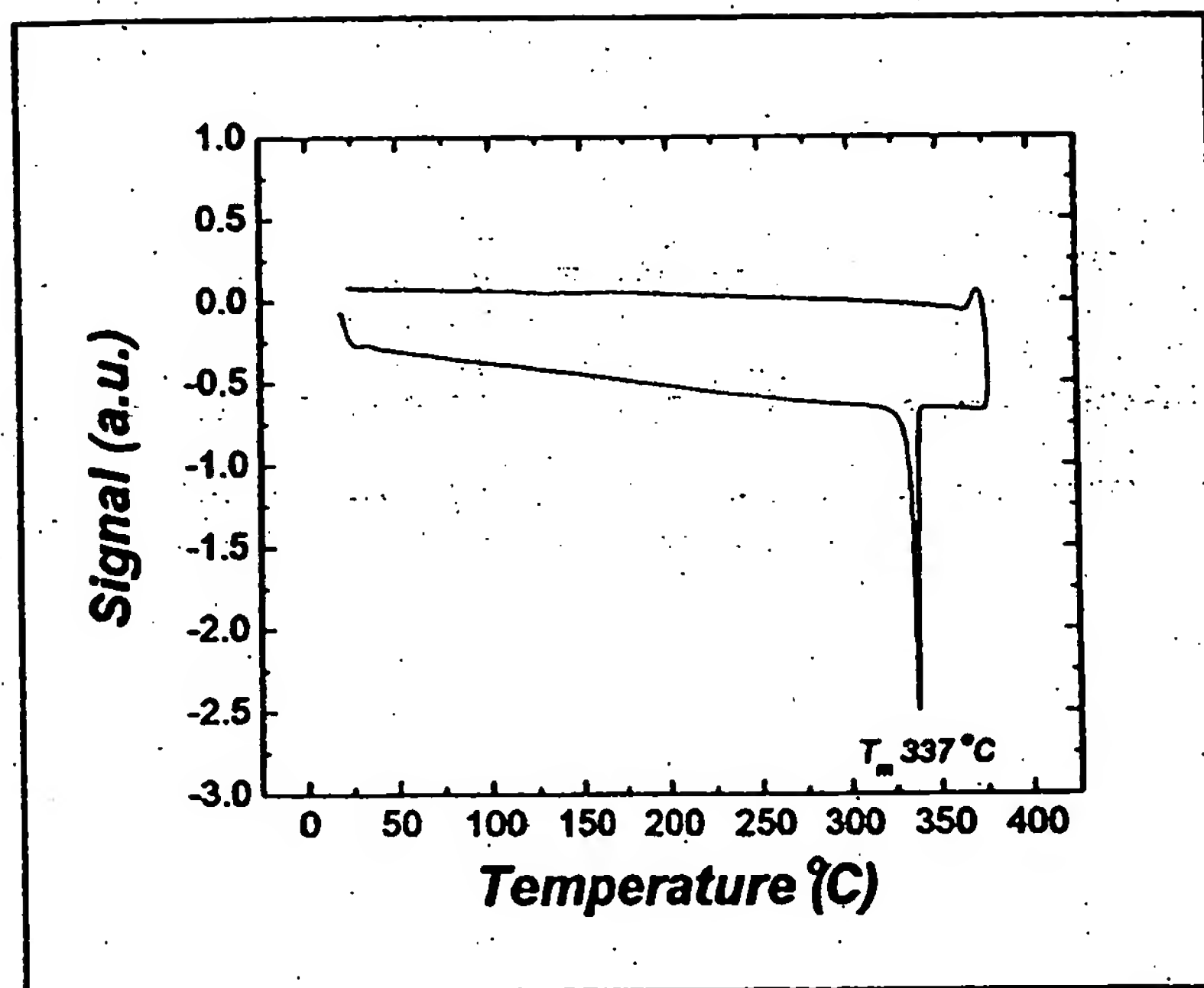
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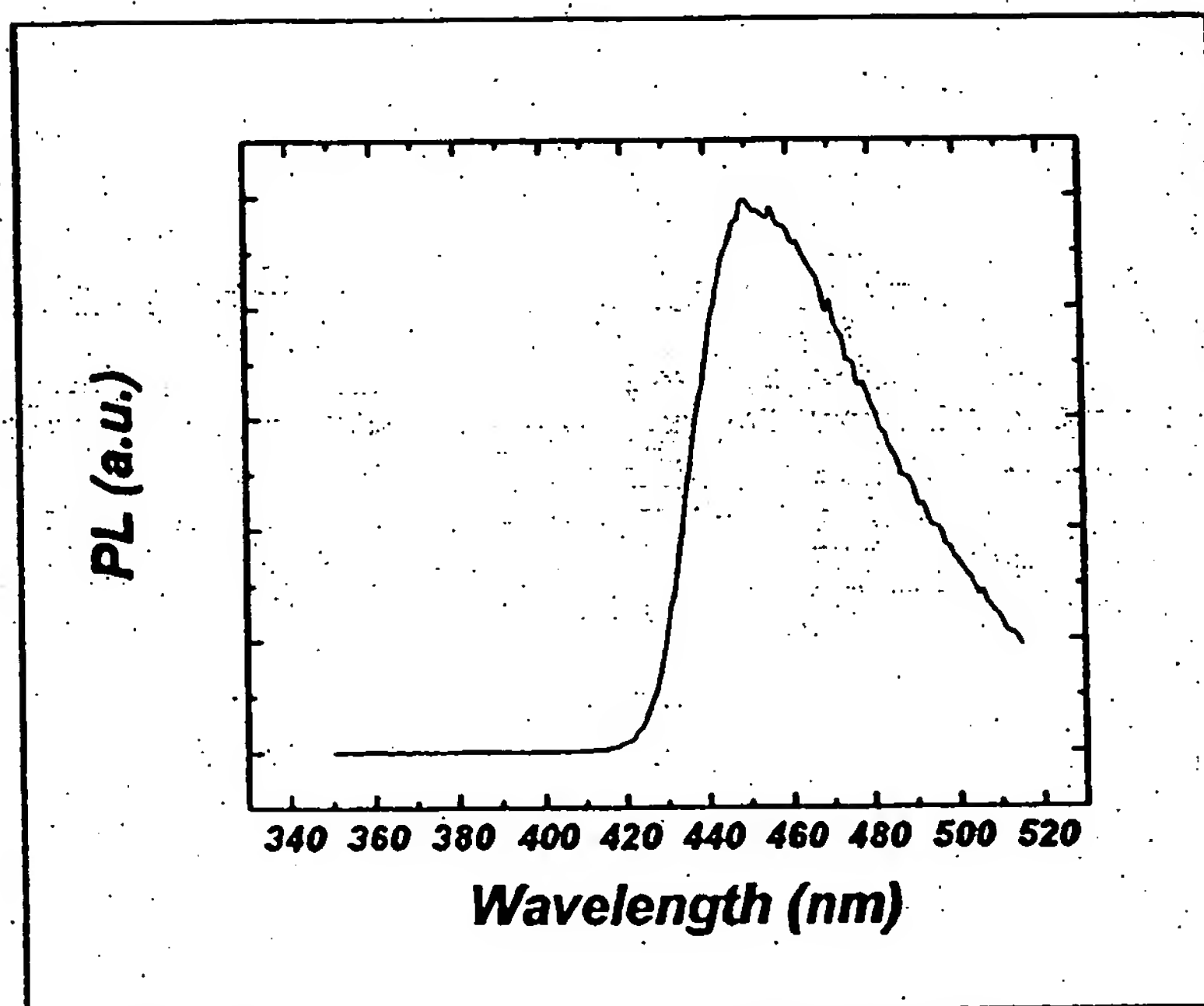
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Figure 1



DSC curve of compound (2).

Figure 2



PL spectrum of compound (4) in CHCl_3 .

FIGURE 3

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EUROPEAN SEARCH REPORT

Application Number
EP 03 25 7582

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| The present search report has been drawn up for all claims | | | |
| Place of search THE HAGUE | | Date of completion of the search 17 March 2004 | Examiner Lehnert, A |
| CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document | | | |



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 03 25 7582

| DOCUMENTS CONSIDERED TO BE RELEVANT | | | |
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| The present search report has been drawn up for all claims | | | |
| Place of search THE HAGUE | | Date of completion of the search 17 March 2004 | Examiner Lehnert, A |
| <p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p> | | | |



European Patent
Office

Application Number

EP 03 25 7582

CLAIMS INCURRING FEES

The present European patent application comprised at the time of filing more than ten claims.

- ☐ Only part of the claims have been paid within the prescribed time limit. The present European search report has been drawn up for the first ten claims and for those claims for which claims fees have been paid, namely claim(s):
- ☐ No claims fees have been paid within the prescribed time limit. The present European search report has been drawn up for the first ten claims.

LACK OF UNITY OF INVENTION

The Search Division considers that the present European patent application does not comply with the requirements of unity of invention and relates to several inventions or groups of inventions, namely:

see sheet 8

- ☐ All further search fees have been paid within the fixed time limit. The present European search report has been drawn up for all claims.
- ☒ As all searchable claims could be searched without effort justifying an additional fee, the Search Division did not invite payment of any additional fee.
- ☐ Only part of the further search fees have been paid within the fixed time limit. The present European search report has been drawn up for those parts of the European patent application which relate to the inventions in respect of which search fees have been paid, namely claims:
- ☐ None of the further search fees have been paid within the fixed time limit. The present European search report has been drawn up for those parts of the European patent application which relate to the invention first mentioned in the claims, namely claims:



European Patent
Office

**LACK OF UNITY OF INVENTION
SHEET B**

Application Number

EP 03 25 7582

The Search Division considers that the present European patent application does not comply with the requirements of unity of invention and relates to several inventions or groups of inventions, namely:

1. Claims: 1-7

An organic light emitting device comprising:
an emissive layer, wherein the emissive layer includes a
binaphthalene or biquinoline derivate according to the
general formula as described in claim 1.

2. Claims: 8-14

An organic light emitting device comprising:
an emissive layer and at least one charge transport layer,
wherein the charge transport layer includes a binaphthalene
or biquinoline derivate according to the general formula as
described in claim 8.

**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 03 25 7582

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report.
The members are as contained in the European Patent Office EDP file on
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

17-03-2004

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